

PCTWORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : C08J 3/00, 3/20, C08L 1/00, 99/00, C08K 11/00, A21C 1/00, B01F 7/00, 11/00		A1	(11) International Publication Number: WO 99/23138 (43) International Publication Date: 14 May 1999 (14.05.99)
(21) International Application Number: PCT/US98/22661 (22) International Filing Date: 26 October 1998 (26.10.98) (30) Priority Data: 08/961,863 31 October 1997 (31.10.97) US (63) Related by Continuation (CON) or Continuation-in-Part (CIP) to Earlier Application US 08/961,863 (CON) Filed on 31 October 1997 (31.10.97) (71) Applicant (for all designated States except US): XYLECO, INC. [US/US]; P.O. Box 36, Brookline, MA 02146 (US). (72) Inventors; and (75) Inventors/Applicants (for US only): MEDOFF, Marshall [US/US]; 90 Addington Road, Brookline, MA 02146 (US). LAGACE, Arthur [US/US]; 21 Prospect Park, Newtonville, MA 02160 (US). (74) Agent: NABINGER, Robert, C.; Fish & Richardson P.C., 225 Franklin Street, Boston, MA 02110-2804 (US).			(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, HR, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG). Published <i>With international search report.</i>
(54) Title: CELLULOSIC FIBER COMPOSITES			
(57) Abstract Composites of a resin and texturized cellulosic or lignocellulosic fiber, and methods for forming the composites, are disclosed.			

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece	ML	Mali	TR	Turkey
BG	Bulgaria	HU	Hungary	MN	Mongolia	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MR	Mauritania	UA	Ukraine
BR	Brazil	IL	Israel	MW	Malawi	UG	Uganda
BY	Belarus	IS	Iceland	MX	Mexico	US	United States of America
CA	Canada	IT	Italy	NE	Niger	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NL	Netherlands	VN	Viet Nam
CG	Congo	KE	Kenya	NO	Norway	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NZ	New Zealand	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	PL	Poland		
CM	Cameroon	KR	Republic of Korea	PT	Portugal		
CN	China	KZ	Kazakhstan	RO	Romania		
CU	Cuba	LC	Saint Lucia	RU	Russian Federation		
CZ	Czech Republic	LI	Liechtenstein	SD	Sudan		
DE	Germany	LK	Sri Lanka	SE	Sweden		
DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

- 1 -

CELLULOSIC FIBER COMPOSITESBackground of the Invention

The invention relates to composites of resins and cellulosic or lignocellulosic fibers.

5 Resins are used in a variety of applications, for example, in food packaging. Food containers made of resins are typically used once, then discarded.

Cellulosic and lignocellulosic materials are produced, processed, and used in large quantities in a
10 number of applications. Once used, these fibers are usually discarded as waste materials. As a result, there is an ever-increasing amount of waste cellulosic and lignocellulosic fiber, as well as waste resin.

Summary of the Invention

15 In general, the invention features composites including a resin and texturized cellulosic or lignocellulosic fiber.

The invention features a composite including a resin, such as a thermoplastic resin, and at least about
20 2% by weight, more preferably at least about 5% by weight, texturized cellulosic or lignocellulosic fiber. The invention also features a composite that includes polyethylene and at least about 50% by weight texturized cellulosic or lignocellulosic fiber.

25 The invention further features composites, including a resin and cellulosic or lignocellulosic fiber, that have flexural strengths of at least about 3,000 psi, or tensile strengths of at least about 3,000 psi.

30 In addition, the invention features a process for manufacturing a composite; the process includes shearing cellulosic or lignocellulosic fiber to form texturized cellulosic or lignocellulosic fiber, then combining the texturized fiber with a resin. A preferred method

- 2 -

includes shearing the fiber with a rotary knife cutter. The invention also features a process for manufacturing a composite that includes shearing cellulosic or lignocellulosic fiber and combining the fiber with a
5 resin.

The term "texturized cellulosic or lignocellulosic fiber" as used herein, means that the fiber has been sheared to the extent that the internal fibers are substantially exposed. At least about 50%, more
10 preferably at least about 70%, of these fibers have a length/diameter (L/D) ratio of at least 5, more preferably at least 25, or at least 50. An example of texturized fiber is shown in Fig. 1.

The composites of the present invention are
15 strong, light-weight, and inexpensive. The raw materials used to make the composites are readily available; for example, they may include discarded containers composed of resins, and waste cellulosic or lignocellulosic fiber.

Other features and advantages of the invention
20 will be apparent from the description of the preferred embodiments thereof, and from the claims.

Brief Description of the Drawing

Fig. 1 is a photograph of a texturized newspaper, magnified [____] times.

Description of the Preferred Embodiments

25 A preferred composite includes a resin and texturized cellulosic or lignocellulosic fiber.

The resin encapsulates the texturized cellulosic or lignocellulosic fiber and helps control the shape of
30 the composite. The resin also transfers the external loads to the fiber and protects the fiber from environmental and structural damage. Preferred composites include about 20% to about 60%, more

- 3 -

preferably about 30% to about 50%, by weight of the resin.

Examples of resins include polyethylene (including, e.g., low density polyethylene and high density polyethylene), polypropylene, polystyrene, polycarbonate, polybutylene, thermoplastic polyesters, polyethers, thermoplastic polyurethane, PVC, Nylon, and other resins. It is preferred that the resins have a low melt flow index. Preferred resins include polyethylene and polypropylene with melt flow indices of less than 3 g/10 min, and more preferably less than 1 g/10 min.

The resins may be purchased as virgin material, or obtained as scrap or waste materials, and are usually purchased in pelletized form. Preferably, the resins are obtained as scrap or waste resins, as these materials are less expensive. A preferred source of resin is used polyethylene milk bottles.

The texturized cellulosic or lignocellulosic fiber provides the composite with strength. The composite may include from about 30% to about 90%, more preferably from about 50% to about 70%, of the texturized cellulosic or lignocellulosic fiber by weight. Examples of cellulosic fiber include paper and paper products; examples of lignocellulosic fiber include wood, wood fibers, and wood-related materials, as well as materials derived from kenaf, grasses, rice hulls, bagasse, cotton, and jute. A preferred cellulosic fiber is newsprint. Preferred lignocellulosic fibers include jute and kenaf.

The composites also include coupling agents. The coupling agents help to bond the hydrophilic fibers to the hydrophobic resins. Examples of coupling agents include maleic anhydride modified polyethylenes, such as those in the FUSABOND® (available from Dupont, Delaware) and POLYBOND® (available from Uniroyal Chemical, Connecticut) series. A preferred coupling agent is a

- 4 -

maleic anhydride modified high density polyethylene such as FUSABOND® MB 100D.

The composites can also contain additives known to those in the art of compounding, such as plasticizers, lubricants, antioxidants, opacifiers, heat stabilizers, colorants, flame retardants, biocides, impact modifiers, photostabilizers, and antistatic agents.

Preparation of Starting Materials

If scrap cellulosic or lignocellulosic fiber is used, it should be cleaned and dried. The cellulosic or lignocellulosic fiber must then be texturized before it is combined with the thermoplastic resin. The fiber can be texturized using any one of a number of mechanical means, or combinations thereof. A preferred method of texturizing includes first cutting the cellulosic or lignocellulosic fiber into 1/4- to 1/2-inch pieces using a standard cutting apparatus. These pieces are then sheared with a rotary cutter, such as the one (available from Sprout, Waldron Companies) described in Perry's Chem. Eng. Handbook, 6th Ed., at 8-29 (1984). The texturized fiber is then passed through a 2 mm mesh screen. It can be stored in sealed bags; it should be dried at approximately 105°C for 4-18 hours (until the moisture content is less than about 0.5%) immediately before use.

The resin may be purchased in a pelletized or granulated form and used without further purification or drying. If surface moisture is present on the pelletized or granulated resin, however, it should be dried before use.

- 5 -

Preparation of Composites

The composites can be prepared as follows. A standard rubber/plastic compounding 2-roll mill is heated to 325-400°C. The resin (usually in the form of pellets or granules) is added to the heated roll mill. After about 10 minutes, the coupling agent is added to the roll mill. After another five minutes, the texturized cellulosic or lignocellulosic fiber is added to the molten resin/coupling agent mixture. The texturized fiber is added over a period of about 10 minutes.

The composite is removed from the roll mill, cut into sheets and allowed to cool to room temperature. It is then compression molded into plaques using standard compression molding techniques.

Alternatively, a mixer, such as a Banbury internal mixer, is charged with the ingredients. The ingredients are mixed, while the temperature is maintained at less than about 190°C. The mixture can then be compression molded.

In another embodiment, the ingredients can be mixed in an extruder mixer, such as a MARIS (Turin) TM 85 extruder equipped with co-rotating screws. The resin and the coupling agent are introduced at the extruder feed throat; the cellulosic or lignocellulosic fiber is introduced about 1/3 of the way down the length of the extruder into the molten resin. The internal temperature of the extruder is maintained at less than about 190°C. At the output, the composite is pelletized by cold strand cutting.

Alternatively, the mixture can first be prepared in a mixer, then transferred to an extruder for the extrusion and pellet-cutting steps.

In another embodiment, the composite can be formed into fibers, using fiber-forming techniques known to those in the art.

- 6 -

Properties of the Composite

The resulting composites include a network of fibers, encapsulated within a resin matrix. The fibers form a lattice network, which provides the composite with strength. Since the cellulosic or lignocellulosic fiber is texturized, the amount of surface area available to bond to the resin is increased, in comparison to composites prepared with un-texturized cellulosic or lignocellulosic fiber. The resin binds to the surfaces of the exposed fibers, creating an intimate blend of the fiber network and the resin matrix. The intimate blending of the fibers and the resin matrix further strengthens the composites.

Uses

The resin/fiber composites can be used in a number of applications. The composites are strong and light weight; they can be used, for example, as wood substitutes. The resin coating renders the composites water-resistant, so they may be used in outdoor applications. For example, the composites may be used to make pallets which are stored outdoors for extended periods of time.

Examples

Composites were prepared as follows. A standard rubber/plastic compounding 2-roll mill was heated to 325-400°C. The resin (usually in the form of pellets or granules) was added to the heated roll mill. After about 10 minutes, the resin banded on the rolls (i.e., it melted and fused to the rolls). The coupling agent was then added to the roll mill. After another five minutes, the cellulosic or lignocellulosic fiber was added to the molten resin/coupling agent mixture. The cellulosic or

- 7 -

lignocellulosic fiber was added over a period of about 10 minutes.

The composite was then removed from the roll mill, cut into sheets, and allowed to cool to room temperature. 5 Batches of about 80 g each were compression molded into 6" x 6" x 1/8" plaques using standard compression molding techniques.

One composition contains the following ingredients:

10	<u>Composition No. 1</u>	
	<u>Ingredient</u>	<u>Amount (g)</u>
	High density polyethylene ¹	160
	Old newspaper ²	240
	Coupling agent ³	8
15	The properties of Composition No. 1 are as follows:	
	Flexural strength (psi)	9,810 (ASTM
	D790)	
	Flexural modulus (10 ⁹ psi)	6.27 (ASTM
	D790)	

20 A second composition contains the following ingredients:

25	<u>Composition No. 2</u>	
	<u>Ingredient</u>	<u>Amount (g)</u>
	High density polyethylene ¹	160
	Old magazines ²	240
	Coupling agent ³	8
	The properties of Composition No. 2 are as follows:	
	Flexural strength (psi)	9,060 (ASTM
	D790)	
30	Flexural modulus (10 ⁹ psi)	6.78 (ASTM
	D790)	

¹ Marlex 16007

² Texturized using rotary cutter with 2 mm mesh

³ FUSABOND® 100D

- 8 -

Other embodiments are within the claims.

What is claimed is:

- 9 -

1. A composite comprising a resin and fiber, wherein the fiber is a cellulosic or lignocellulosic fiber, and at least about 2% by weight of the fiber is texturized.
- 5 2. The composite of claim 1, wherein at least about 5% by weight of the fiber is texturized.
3. The composite of claim 1, wherein the fiber is newsprint.
4. The composite of claim 1, wherein the fiber is
10 jute.
5. The composite of claim 1, wherein the fiber is kenaf.
6. The composite of claim 1, wherein the resin is magazine paper.
- 15 7. The composite of claim 1, wherein the resin is beached craft board.
8. The composite of claim 1, wherein the resin is a thermoplastic resin.
9. The composite of claim 6, wherein the
20 thermoplastic resin is polyethylene.
10. The composite of claim 6, wherein the thermoplastic resin is polypropylene.
11. The composite of claim 1, wherein the composite comprises about 50% to about 70% by weight
25 resin and about 37% to about 50% by weight fiber.

- 10 -

12. A composite comprising polyethylene and fiber, wherein the fiber is cellulosic or lignocellulosic fiber, and wherein at least about 50% by weight of the fiber is texturized.

5 13. A composite comprising a resin and cellulosic or lignocellulosic fiber, wherein the composite has a flexural strength of at least 3,000 psi.

14. The composite of claim 11, wherein the composite has a flexural strength of at least 6,000 psi.

10 15. The composite of claim 11, wherein the composite has a flexural strength of at least 10,000 psi.

16. A process for manufacturing a composite comprising shearing cellulosic or lignocellulosic fiber to form texturized cellulosic or lignocellulosic fiber,
15 and combining the texturized cellulosic or lignocellulosic fiber with a resin.

17. The process of claim 16, wherein the resin is a thermoplastic resin.

18. The process of claim 17, wherein the step of
20 shearing the cellulosic or lignocellulosic fiber comprises shearing with a rotary knife cutter.

19. A process for manufacturing a composite, the process comprising shearing cellulosic or lignocellulosic fiber and combining the cellulosic or lignocellulosic
25 fiber with a resin.

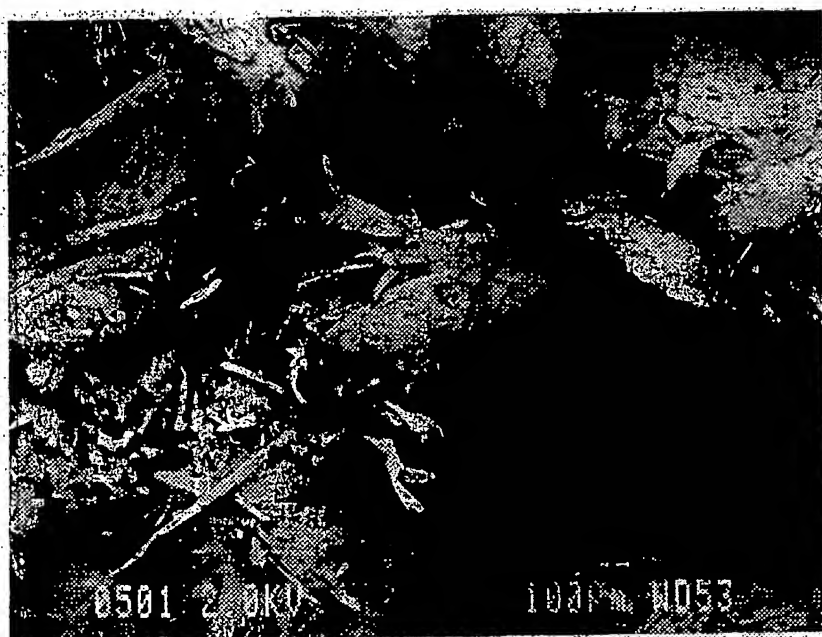


FIG. 1

INTERNATIONAL SEARCH REPORT

 International application No.
 PCT/US98/22661

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : Please See Extra Sheet.

US CL : 523/129; 524/13, 14, 35, 72, 76; 366/69, 241

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 523/129; 524/13, 14, 35, 72, 76; 366/69, 241

 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
 NONE

 Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
 NONE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X -- Y	US 4,791,020 A (KOTKA) 13 December 1988, see col. 3, lines 17-20 and col. 3, line 41, to col. 4, line 8.	1-9, 11, 12 ----- 1-15
X	US 5,516,472 A (LAVER) 14 May 1996, see col. 1, lines 27-44, col. 6, lines 29-64, and col. 8, lines 18-29.	1-3, 5, 6, 8-11
X	US 4,244,847 A (POSIVIATA et al) 13 January 1981, see col. 2, lines 42 et seq, and col. 4, lines 32 et seq.	1-4, 6-8, 11, 12

☐ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

* Special categories of cited documents:	*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
A document defining the general state of the art which is not considered to be of particular relevance	*X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
B earlier document published on or after the international filing date	*Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	*Z* document member of the same patent family
O document referring to an oral disclosure, use, exhibition or other means	
P document published prior to the international filing date but later than the priority date claimed	

Date of the actual completion of the international search

19 DECEMBER 1998

Date of mailing of the international search report

28 JAN 1999

 Name and mailing address of the ISA/US
 Commissioner of Patents and Trademarks
 Box PCT
 Washington, D.C. 20231

Facsimile No. (703) 305-3230

Authorized officer

NATHAN M. NUTTER

Telephone No. (703) 308-0561

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US98/22661

A. CLASSIFICATION OF SUBJECT MATTER:

IPC (6):

C08J 3/00, 3/20; C08L 1/00, 99/00; C08K 11/00; A21C 1/00; B01F 7/00, 11/00